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Effects of Material Viscosity on Particle Sizing by Ultrasonic Attenuation Spectroscopy

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Abstract

Ultrasonic spectroscopy is becoming a promising particle measurement technique for the characterization of emulsions and suspensions over a wide range of particle size and concentration especially in the petrochemical, pharmaceutical, adhesives, food and other industries. Compared with other competitive methods like laser diffraction, dynamic light scattering and image analysis, the approach of ultrasonic attenuation spectroscopy possesses significant superiority in the highly-concentrated particulate two-phase flow measurement even in the optical opaque medium, due to the high penetrability of ultrasonic waves through medium, as well as in situ non-invasive measurement and low cost but endurable apparatus. Typically, the overall process by which the particle size distribution of a suspension is measured using ultrasonic attenuation spectroscopy can be divided into two parts: the measurement of frequency-dependent ultrasonic attenuation in two-phase system, and the inversion calculation based on the prediction of mathematical model. The modeling process relies on the fundamental relationship between attenuation spectrum and physical properties, where viscosity could be a critical and complex property because its quantity can influence the ultrasonic attenuation seriously and ultimately the inversion calculation of the resultant particle size distribution. In this paper, a variable viscosity experimental system has been established through changing the concentration of aqueous glycerol solutions or the temperature of glycerol solutions. For the sample of micron-sized glass beads, a series of experiments have been carried out to obtain the attenuation spectrum, which were predicted and interpreted using the ECAH (Epstein, Carhart, Allegra and Hawley) model simultaneously. Hence, the particle size distribution of the glass beads can be retrieved through the inversion calculation.

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Keywords: Ultrasonic; Attenuation spectrum; Viscosity; Particle size distribution

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1. Introduction

In recent years, ultrasound technique has gained considerable attention for particle measurement, offering advantages with respect to more extensively accepted analytical methods such as optical and electrical techniques. Characteristics like the availability to optically opaque, undiluted and high/non-conducting dispersions and the non-intrusive nature of the acoustic transducers promote its applications for the rapidly and on-line monitoring of a wide field such as petrochemical, pharmaceutical, and food industries [1].

Furthermore, ultrasound attenuation spectroscopy has been validated to be applicable to the characterization of particles over several orders of magnitude in size (10 nm to 1 mm) at very high concentrations (volume fraction from 0.1% to 50%) [2]. By making use of a rigorous and fundamental theoretical model, particle sizing with ultrasound spectroscopy deeply relies on the assumption that the fundamental relationship between attenuation spectrum and requisite physical properties, where viscosity could usually be a significant and complicated property because its quantity can influence the ultrasonic attenuation seriously by direct sound absorption of continue media and viscos loss occurred in the interaction between sound and media, and ultimately the inversion calculation of the resultant particle size distribution. While, Frank Babick *et al* focused on the dependence of ultrasonic attenuation on the material properties and illuminated which material properties have to be obtained with high accuracy and which can be estimated [3]. As Patricia Mougina *et al* have studied the sensitivity of particle sizing by ultrasonic attenuation spectroscopy to material properties [4].

In this study, we investigate the influence of viscosity on the ultrasonic attenuation coefficient and particle size distribution of micron-sized glass beads in glycerol solutions or aqueous glycerol solutions with different temperature and volume concentration, respectively. We also consider the implications of the viscosity-dependence of the ultrasonic properties of suspensions for utilization of the ultrasound technique for characterizing suspensions. In Section 2, a brief introduction to the physical background of ultrasound technique as well as associated fundamental model are presented. Then in Section 3, a simple presentation to the materials and experiments and an analytical derivation of the dependence of obtained measured and calculated results from viscosity are given. After that, the analytical results are verified and discussed by calculating particle size distributions of measured attenuation spectrum with different sets of material properties changing with viscosity in Section 4, followed by a final conclusion. In particular, the study aims to characterize and compare dispersions with different volume concentrations and different temperature in order to demonstrate the possible ways in which the change of viscosity of dispersion can change the original ultrasonic attenuation spectrum of the dispersion. The study brings the conditions in which ultrasonic attenuation spectroscopy is capable of explaining that the change of viscosity of a dispersion will change original particle size of the disperse medium.

2. Theoretical basis

2.1. Physical background of ultrasonic attenuation spectroscopy

In a homogeneous medium, sound waves, similar to light, propagate straight from the source, thus steadily losing part of their energy during sound absorption in the dispersed medium. In heterogeneous media, such as in suspensions or emulsions, the redirection of acoustic energy away from the incident beam can be observed in sound propagation [5]. Sound scattering is a general wave phenomenon that the sound waves are refracted, reflected and diffracted on the dispersed phase in such a way that each particle acts as a sound source, radiating sound waves in all directions. Against light scattering, sound scattering could be strongly affected by thermal and mechanical coupling effects between the continuous and dispersed phase. That's the physical characteristic of sound waves, which are nothing less than propagating compressional equilibrium disturbances, consistent to temperature fluctuations. The thermal and mechanical coupling effects impact the sound propagation in two ways: they vary the scattering profile, and they are of a dissipative nature hence decreasing the total amount of acoustic energy through sound absorption. The magnitude of the scattering effect depends on the physical properties of the involved phases and the particle size [6].

2.2. Mathematical modeling of ultrasonic attenuation

When sound waves pass through suspensions, alterations occur to the sound waves as well as to the continuous and dispersed phase. The attenuation of ultrasound waves passing through suspensions can be predicted when a set of thermodynamic, transport and mechanical properties describing both the continuous and dispersed phase is given [7]. Models derived from the scattering theory depict the acoustic behavior of dispersions for all frequencies and particle sizes. A thorough explanation for non-gaseous particles in a liquid continuum is proposed as Epstein-Carhart and Allergr-Hawley models (ECAH) [8, 9], based on the effect of a continuous ultrasound wave scattered on a single spherical particle of a given size. The classical ECAH model has been adopted as a mathematical method to carry out numerical calculations in order to clarify the relationship and sensitivity between ultrasound spectrum and particle size [10]. The relative significance of the different mechanisms of sound absorption through dispersed phase depends upon the frequency of the ultrasound wave, the thermo-physical properties of the two phases in the dispersed system as well as the size and concentration of the particles [11]. As poly-disperse particle system the problem has not been involved in the ECAH model itself [12]. Considering the contribution of all size bins according to a summation, the corresponding formulae can be given by:

$$\alpha_s = \frac{3\phi}{2k^2}\sum_{j=1}^{N}\frac{q_j}{R_j^3}\sum_{n=0}^{\infty}(2n+1)\,\mathrm{Re}(A_n(R_j,\omega)) \tag{1}$$

$$\frac{1}{c_s}-\frac{1}{c_w} = \frac{3\phi}{2\omega k^2}\sum_{j=1}^{N}\frac{q_j}{R_j^3}\sum_{n=0}^{\infty}(2n+1)\,\mathrm{Im}(A_n(R_j,\omega)) \tag{2}$$

For ultrasonic attenuation coefficient α_s and phase velocity C_s of the two-phase flow: $\emptyset$ is the disperse volume fraction; k is the complex propagation constant of the incident compression wave; q_j is the volume fraction of particle radius in the j-th size range $[R_j, R_{j+1}]$; R indicates the particle radius; A_n is the scattering coefficient, which is the key for calculating ultrasonic attenuation coefficient α_s and phase velocity C_s; $\omega=2\pi f$ is the angular frequency; f is the frequency; C_w is the sound velocity of water.

3. Materials and methods

3.1. Materials and sample preparation

Experiments were conducted using micron-sized glass beads. The density, weight median particle diameter and index of refraction of small glass beads are 4.19 g/cm^3, 19.3±1.1 μm and 1.90(@589nm), separately. There were two groups of solution samples: one group of solution samples was prepared with a mixture of distilled water and glycerin at 20℃, in which the volume fraction of aqueous glycerol solutions were 50%, 60%, 70%, 80%, 90% and 100%, respectively; the other group of samples was prepared by controlling the temperature of glycerol solutions to 20℃, 30℃, 35℃, 45℃ and 55℃, respectively. Then, glass beads as dispersed particles and these solutions as continuous media were separately mixed and the volume ratio of particles to solutions was kept at 1%. The path-length through samples was then precisely determined by the thickness of the sample cells (l_1=10mm or l_2=20mm).

3.2. Ultrasonic experiments

As shown in Fig.1, a typical water-immersed ultrasonic experimental system was used for a through-transmission mode measurement, where two fixed broadband transducers T1 & T2, (Panametrics M310-SU, the nominal frequency: 5MHz) served as a transmitter and a receiver. A transmitting & receiving ultrasonic signal analyzer (Panametrics 5800PR) excited the transmitter with a sinusoidal burst wave. Ultrasonic longitudinal wave propagated in water-sample-water and was received by the receiver. After converted by the A/D (NI PCI-5114), the signal was

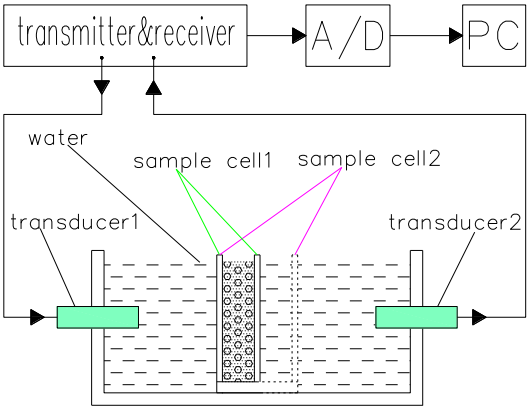


Fig.1. Schematic representation of experimental system

processed into amplitude (A_1 or A_2, $A_1 > A_2$) through the fast Fourier transform. Finally, the wave attenuation in the sample α_l was obtained by comparing the wave amplitudes of samples with different thickness:

$$\alpha_l = \frac{\ln(A_1 / A_2)}{l_2 - l_1} + \alpha_w \tag{3}$$

where α_w is the wave attenuation in water, A_1 denotes the amplitude corresponding to the sample thickness of l_1(10mm) and A_2 denotes the amplitude corresponding to the sample thickness of l_2(20mm). Additionally, a series of ultrasonic procedures converting the ultrasound signals into particle size distribution has been compiled. A modified version of the nonlinear iterative Chahine algorithm was used in the inversion calculation of particle size [13]. Compared with the algorithm originally proposed by Chahine, this method is much more stable with respect to random noise, permits a better quality of the retrieved distributions, and improves the overall reliability of the fitting [14].

4. Results and discussion

The viscosity of aqueous glycerol solutions at 50%, 60%, 70%, 80%, 90% and 100% volume fraction measured by a commercial viscometer(SOPTOP SNB-1) at 20℃ were 6.05 mPa•s, 10.96 mPa•s, 22.94 mPa•s, 62 mPa•s, 234.6 mPa•s and 1499 mPa•s, respectively. While, the viscosity of glycerol solutions at 20℃, 30℃, 35℃, 45℃ and 55℃ were 1499 mPa•s, 624 mPa•s, 448 mPa•s, 213 mPa•s and 111.65 mPa•s, separately. And, the viscosity of distilled water at 20℃ were 0.903 mPa•s.

4.1. Measurement results at varying volume concentrations

Fig.2 illustrates the frequency-dependent ultrasonic attenuation in the glass particles compounded aqueous glycerol solutions of different volume fraction. As seen in Fig.2, ultrasonic attenuation raised with the increasing volume concentration of aqueous glycerol solutions since the viscosity increased as the increasing volume concentration significantly, from 6.05 mPa•s to 1499 mPa•s. This is much reasonable considering the frictional and scattering attenuation processes [15]. Fig.3 illuminates the particle size distribution of glass beads in water and aqueous glycerol solutions at different volume fraction. There are notable differences among these measured results, but not an obviously increasing or decreasing trend. These differences are extremely probably caused by the viscosity variation because the viscosity of this group samples varied obviously with respect to other physical properties needed in the numerical simulation.

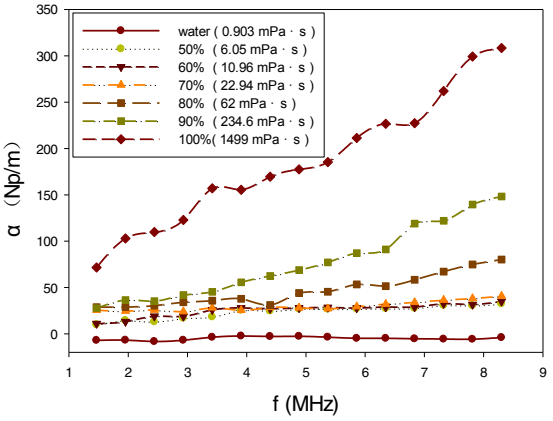


Fig.2. Measured attenuation spectrum of glass beads in the water and aqueous glycerol solutions with different volume concentration at 20℃.

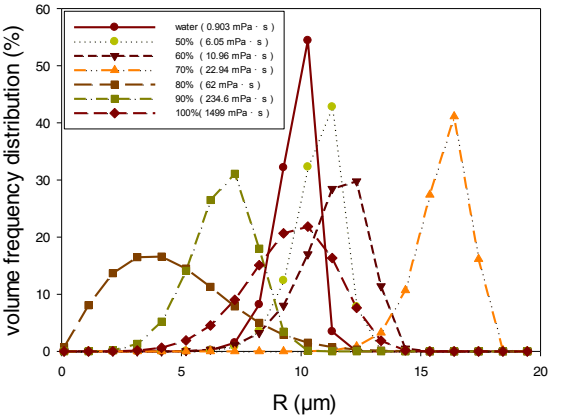
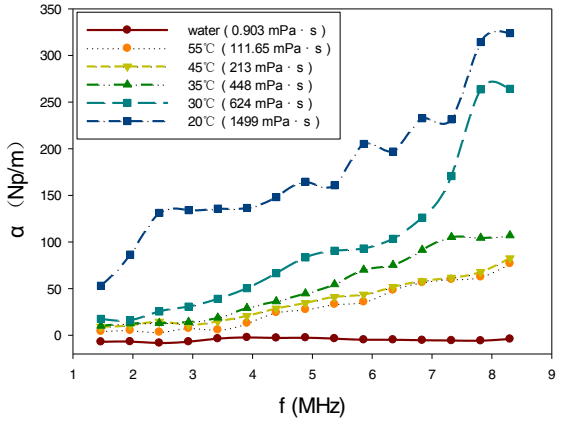


Fig.3. Measured size distribution of glass beads in the water and aqueous glycerol solutions with different volume concentration at 20℃.

4.2. Measurement results at varying temperatures

The ultrasonic attenuation spectrum in the glass particles compounded glycerol solutions of different temperature are shown in Fig.4. As can be seen in Fig.4, attenuation reduced with the increasing temperature of glycerol solutions that the viscosity decreased as the increasing temperature signally, from 111.65 mPa•s to 1499 mPa•s. This is also reasonable considering the frictional and scattering attenuation processes. Fig.5 illustrates the particle size distribution of water and glycerol solutions at different temperatures. There are significantly differences among these measured results without the law of increase or decrease, either. It can be inferred that the differences are partly caused by the viscosity change since the viscosity of this group samples also varied distinctly with respect to other physical properties needed in the numerical calculation.

Certainly, there are some other varying physical properties, such as density, sound velocity, sound absorption, temperature and thermal conductivity. In order to reveal the dominant physical properties causing the difference of particle size distribution and evaluate the effect of viscosity, a series of numerical simulation were implemented, investigating the effect of density, sound velocity, sound absorption, temperature and thermal conductivity on particle size distribution.



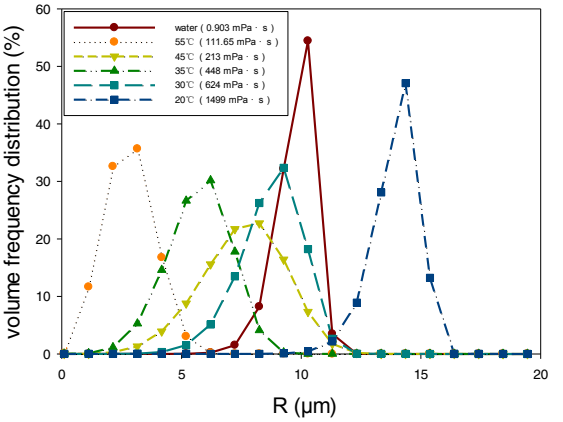


Fig.4. Measured attenuation spectrum of glass beads in the water and glycerol solutions at different temperatures.

Fig.5. Measured size distribution of glass beads in the water and glycerol solutions at different temperatures.

5. Simulation results and analysis

The reference values of physical properties for the experimental systems are listed in Table.1 and Table.2. These physical properties were either measured directly or obtained from the literature. It is shown that there are some other varying physical properties except viscosity, such as density, sound velocity, sound absorption, temperature and thermal conductivity. The maximal variations of density, sound velocity, sound absorption, temperature and thermal conductivity are severally 10.71%, 11.59%, 86.93%, 10.67% and 31.32% calculated by the values of Table.1 and Table.2. As a result, these physical properties in turn were modified by ±11%, ±12%, ±87%, ±11% and ±12%, respectively. And, the calculated numerical simulated results of alteration of these physical properties were compared by the original parameters of particle-in-glycerol solutions-suspension at 30℃(Fig.6-Fig.10). As can be seen in Fig.6, it generates a slight variation on the particle size distribution altering the density by +11%, but scarcely any influence by -11% (Fig.6). Contrary to this, it generates a slight variation on the particle size distribution altering the sound speed by -12%, but scarcely any influence by +12% (Fig.7). Relatively, it generates greater variation on the particle size distribution altering the sound absorption by -87%, but hardly any influence by +87% (Fig.8). Nevertheless, it generates barely any effect on the particle size distribution altering the temperature and thermal conductivity by ±11% and ±32% (Fig.9 and Fig.10). Subsequently, a refined analysis was performed which demonstrated the effect of alteration of material properties on the calculated characteristic particle size and distribution width from the measured attenuation spectrum (Table.3). The values of variations of the number median diameter D_{N50}, volume median diameter D_{V50}, Sauter mean diameter D_{32} and distribution width Dw_1/ Dw_2 were all less than 2.1%. However, the value of diversities ranged from -38.5% to 30.6% by calculating the Sauter mean diameters of two group solution samples from the data in Fig.3 and Fig.5. Thus, as it were, it is viscosity predominantly causing the significant difference of inversed glass particle size distributions.

Table 1. Reference physical properties of water and aqueous glycerol solutions with different volume concentration at 20℃.

Property	Density ($kg•m^{-3}$)	Sound speed ($m•s^{-1}$)	Viscosity (Pa•s)	Sound absorption ($Np•m^{-1}•Hz^{-2}$)	Temperature (K)	Thermal expansion (K^{-1})	Specific heat ($J•kg^{-1}•K^{-1}$)	Thermal conductivity ($W•m^{-1}•K^{-1}$)
water	997.04	1496.70	0.000903	$2.2*10^{-14}$	293.15	0.000257	4178.5	0.5952
50%	1122.37	1679.90	0.006050	$10.5*10^{-14}$	293.15	0.000490	2427.8	0.3564
60%	1149.29	1723.50	0.010960	$11.4*10^{-14}$	293.15	0.000490	2427.8	0.3276
70%	1178.29	1777.00	0.022940	$12.7*10^{-14}$	293.15	0.000490	2427.8	0.3024
80%	1204.11	1831.45	0.062000	$19.8*10^{-14}$	293.15	0.000490	2427.8	0.2808
90%	1231.02	1875.10	0.234600	$32.9*10^{-14}$	293.15	0.000490	2427.8	0.2592
100%	1256.94	1900.00	1.499000	$80.3*10^{-14}$	293.15	0.000490	2427.8	0.2448

Table 2. Reference physical properties of water and glycerol solutions at different temperatures.

Property	Density ($kg•m^{-3}$)	Sound speed ($m•s^{-1}$)	Viscosity (Pa•s)	Sound absorption ($Np•m^{-1}•Hz^{-2}$)	Temperature (K)	Thermal expansion (K^{-1})	Specific heat ($J•kg^{-1}•K^{-1}$)	Thermal conductivity ($W•m^{-1}•K^{-1}$)
water	997.04	1496.70	0.000903	$2.2*10^{-14}$	293.15	0.000257	4178.5	0.5952
55℃	1235.72	1835.95	0.111650	$13.2*10^{-14}$	328.15	0.000490	2427.8	0.2448
45℃	1241.72	1854.25	0.213000	$15.7*10^{-14}$	318.15	0.000490	2427.8	0.2448
35℃	1247.77	1872.55	0.448000	$22.2*10^{-14}$	308.15	0.000490	2427.8	0.2448

30℃	1250.82	1881.70	0.624000	$40.7*10^{-14}$	303.15	0.000490	2427.8	0.2448
20℃	1256.94	1900.00	1.499000	$75.4*10^{-14}$	293.15	0.000490	2427.8	0.2448

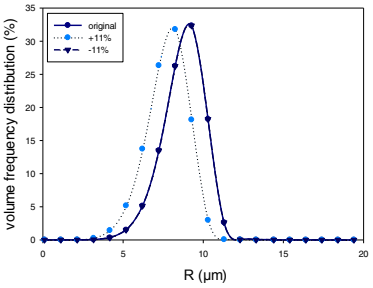


Fig.6 Comparison of the calculated particle size distribution from the measured attenuation spectrum by using the original density and an error superposition by ±11%.

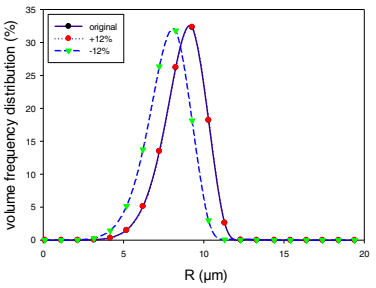


Fig.7 Comparison of the calculated particle size distribution from the measured attenuation spectrum by using the original sound speed and an error superposition by ±12%.

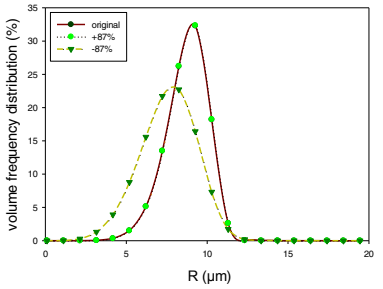


Fig.8 Comparison of the calculated particle size distribution from the measured attenuation spectrum by using the original sound absorption and an superposition by ±87%.

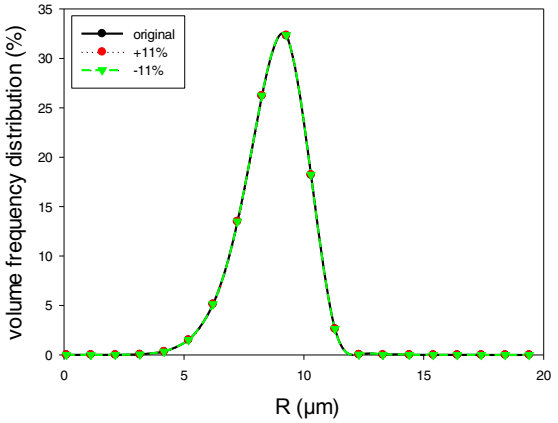


Fig.9 Comparison of the calculated particle size distribution from the measured attenuation spectrum by using the original temperature and an error superposition by ±11%.

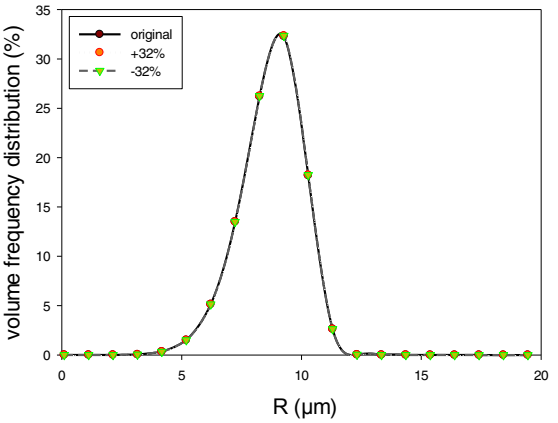


Fig.10 Comparison of the calculated particle size distribution from the measured attenuation spectrum by using the original thermal conductivity and an error superposition by ±32%.

Table 3. Effect of alteration of material properties on the calculated characteristic particle size and distribution width.

Parametric variation		D_{N50} (μm)	D_{V50} (μm)	D_{32} (μm)	$DW_1= D_{N90}-D_{N10}$ (μm)	$DW_2= D_{V10}-D_{V90}$ (μm)
Original value		15.48	17.51	17.87	6.11	6.11
Density	+11%	15.48	17.51	17.36	8.15	6.11
	Δ	0	0	-0.51	2.04	0
	-11%	15.48	17.51	17.98	6.11	6.11
	Δ	0	0	0.11	0	0
Sound speed	+12%	17.51	17.51	18.37	6.11	6.11
	Δ	2.03	0	0.5	0	0
	-12%	15.48	15.48	16.7	4.07	6.11
	Δ	0	-2.03	-1.17	-2.04	0
Sound absorption	+87%	15.48	17.51	17.72	6.11	6.11

	Δ	0	0	-0.15	0	0
	-87%	13.44	15.48	16.6	8.14	8.15
	Δ	-2.04	-2.03	-1.27	2.03	2.04
	+11%	15.48	17.51	17.86	6.11	6.11
Temperature	Δ	0	0	-0.01	0	0
	-11%	15.48	17.51	17.87	6.11	6.11
	Δ	0	0	0	0	0
	+32%	15.48	17.51	17.86	6.11	6.11
Thermal conductivity	Δ	0	0	-0.01	0	0
	-32%	15.48	17.51	17.88	6.11	6.11
	Δ	0	0	0.01	0	0

6. Conclusions

In this study, we focused on the effects of material viscosity on particle sizing by ultrasonic attenuation spectroscopy through experiments and numerical simulations. The experimental and simulated results indicate that for the number median diameter, volume median diameter, Sauter mean diameter and distribution width, the values of variations caused by other varying material physical properties except viscosity were all less than 2.1%. Nevertheless, the value of diversities ranged from -38.5% to 30.6% by calculating the Sauter mean diameters of two group solution samples from the data of measured particle size distribution. Thus, it is the viscosity predominantly causing the significant difference of inversed glass particle size distributions.

Acknowledgements

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